The problem with negative heat capacities for nuclei [1]

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Anomalous negative heat capacities have been claimed as indicators of first order phase transitions in finite systems in general, and for nuclear systems in particular. Creation of additional surface in the guise of bubbles, fragments, etc. has been generically considered responsible for such an effect.

In the case of nuclei, we can rely on the Clapeyron equation $(dp/dT = \Delta H_m/T\Delta V_m)$ to calculate what one might reasonably expect for the heat capacity (C_p) at constant pressure as follows:

$$C_p = \left. \frac{dH}{dT} \right|_p = \left. \frac{dH}{dA} \right|_p \left. \frac{dA}{dT} \right|_p = \Delta H_m(A) \frac{dA}{dT} \right|_p \tag{1}$$

where ΔH_m is the molar vaporization enthalpy. Note that

$$\left. \frac{dT}{dA} \right|_{p} = -\left. \frac{dp}{dA} \right|_{T} \left. \frac{dT}{dp} \right|_{A}. \tag{2}$$

From the integrated form of the Clapeyron equation we have

$$\left. \frac{dp}{dA} \right|_T = -\frac{p}{T} \frac{d\Delta H_m}{dA} \tag{3}$$

so

$$\left. \frac{dT}{dA} \right|_{r} = \frac{p}{T} \frac{d\Delta H_m}{dA} \frac{T^2}{p\Delta H_m} = \frac{T}{\Delta H_m} \frac{d\Delta H_m}{dA}. \tag{4}$$

Finally,

$$C_p = \frac{\frac{(\Delta H_m(A))^2}{T}}{\frac{d\Delta H_m}{dA}} \tag{5}$$

The derivative in the denominator can be evaluated approximately from the dependence on the binding energy per nucleon B upon the mass number

$$\frac{d\Delta H_m}{dA} = -\frac{dB}{dA}. (6)$$

The liquid drop model allows us to estimate such a derivative. Without the Coulomb term, we recover the results for a drop: the binding energy increases with increasing A and tends asymptotically to the value $a_v \approx 15 \text{MeV}$. Thus negative heat capacities should be expected. The Coulomb and symmetry terms, however, become very important at large values of A, say, along the line of β -stability. From Fig. 1 it is apparent that

the binding energy decreases with A for $A > \sim 60$. Consequently in all this region of A, positive specific heats

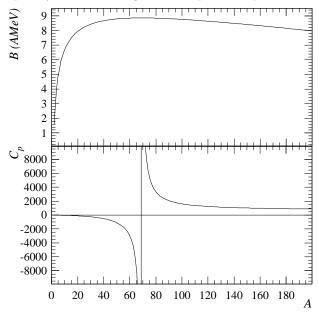


FIG. 1: Top: the binding energy per nucleon of atomic nuclei. Bottom: the associated heat capacity.

should be expected. Only for $A<\sim 60,$ negative specific heats are predicted in this description.

However, the Coulomb force, being long range, creates a much more serious problem of principle in defining statistical and thermodynamic equilibria. Since Coulomb makes a nucleus with $A \approx 200$ metastable by hundreds of MeV, no meaningful statistical treatment is possible, let alone the calculation of caloric curves and heat capacities. In these cases equilibria are only defineable locally, and if this problem can be dealt with at all, the leading effect will be a change in the binding energy.

This straightforward result based on elementary thermodynamics and ground state binding energies raises serious questions as to the meaning of the negative heat capacities that have been claimed for large nuclear systems.

[1] Condensed from Phys. Rev. C 66, 041601(R) (2002).